

# Metal-to-Insulator Crossover in $\text{YBa}_2\text{Cu}_3\text{O}_y$ Probed by Low-Temperature Quasiparticle Heat Transport

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It was recently demonstrated that in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  the magnetic-field ( $H$ ) dependence of the low-temperature thermal conductivity  $\kappa$  up to 16 T reflects whether the normal state is a metal or an insulator. We measure the  $H$  dependence of  $\kappa$  in  $\text{YBa}_2\text{Cu}_3\text{O}_y$  (YBCO) at subkelvin temperatures for a wide doping range, and find that at low doping the  $\kappa(H)$  behavior signifies the change in the ground state in this system as well. Surprisingly, the critical doping is found to be located deeply inside the underdoped region, about the hole doping of 0.07 hole/Cu; this critical doping is apparently related to the stripe correlations as revealed by the in-plane resistivity anisotropy.

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It is well recognized that the high- $T_c$  superconductivity in cuprates is realized in a strongly-correlated electron system that is essentially a doped Mott insulator. Studying the low-temperature normal state, the ground state in the absence of superconductivity, is important in understanding the rather mysterious electronic state from which the superconductivity emerges. In the past, the low-temperature normal state has been investigated by measuring the resistivity while destroying the superconductivity with a very high magnetic field in systems that have relatively low  $T_c$ , such as  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO) [1, 2],  $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+\delta}$  (BSLCO) [3], and  $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$  (PCCO) [4]. In these experiments, it was revealed that the metal-to-insulator crossover (MIC) in the low-temperature normal state takes place at optimum doping (hole doping per Cu,  $p$ , of  $\sim 0.16$ ) in LSCO [2] and in PCCO [4], while it occurs in the underdoped region (specifically, at  $p \sim 1/8$ ) in BSLCO [3]. Thus, although the critical doping for the MIC was found to be nonuniversal, it was established that there is a certain range of doping where the superconductor has an “insulator” as its normal state under high magnetic fields. This insulator is unusual in that it shows a peculiar  $\log(1/T)$  divergence in resistivity [1, 2, 3].

Since it is exotic to have an “insulating” normal state in a superconductor, and furthermore the properties of the insulator are unusual, it is expected that the nature of this insulator bears a key to the microscopic understanding of the electronic state of the cuprates. Given that the critical doping for the MIC is different for LSCO and BSLCO, it is clearly desirable to study the MIC in other cuprate systems and gain insights into the cause of the insulating behavior. Obvious targets for such studies are  $\text{YBa}_2\text{Cu}_3\text{O}_y$  (YBCO) and  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Bi2212), but the upper critical fields of these materials are so high that it has been difficult to observe the MIC at low temperatures.

Recently, we have demonstrated that the low-temperature quasiparticle (QP) heat transport measured in 16-T field can be an alternative tool to probe the MIC

[5]; namely, we found in LSCO that whether the low-temperature normal state under 60 T [2] is metallic or insulating corresponds exactly to whether the QP heat transport at very low temperature is enhanced or suppressed by magnetic fields up to 16 T. (Essentially the same result was reported independently by Hawthorn *et al.* [6].) It was discussed [5] that the magnetic-field suppression of  $\kappa$  at very low temperature is related to the magnetic-field-induced spin density wave (SDW) [7, 8] or charge density wave (CDW) [9], that appear to be competing with the superconductivity and would localize the QPs, thereby leading to the insulating state. It has been proposed [10] that these magnetic-field-induced orders may be related to charge stripes [11], which have been shown to be relevant to the charge transport in cuprates at least in the lightly hole-doped region [12, 13, 14, 15]. In fact, a recent theory showed [16] that the magnetic-field-induced incommensurate antiferromagnetism can be responsible for the localized QP transport at low temperature. In any case, the result in LSCO tells us that the magnetic-field dependence of  $\kappa$  at low-temperatures in YBCO would clarify where the MIC lies in this 90-K-class superconductor. The YBCO system is particularly useful for examining the relevance of the charge-stripe instability, because this is the only system other than the La-based cuprates where the neutron scattering has found static charge stripes [17].

In this Letter, we locate the MIC in YBCO according to the above strategy. We find that the critical doping is unexpectedly low, at the oxygen content  $y$  of  $\sim 6.55$ , which corresponds to  $p \sim 0.07$ . Interestingly, this low critical doping is exactly the same as the boundary below which the charge stripes start to dominate the charge transport, as evidenced by our study of the in-plane resistivity anisotropy in this material [14]. This result gives evidence that the insulating normal state of the cuprate under high magnetic field is related to the self-organization of the electrons, perhaps into the form of disordered stripes.

High-quality  $\text{YBa}_2\text{Cu}_3\text{O}_y$  single crystals are grown in

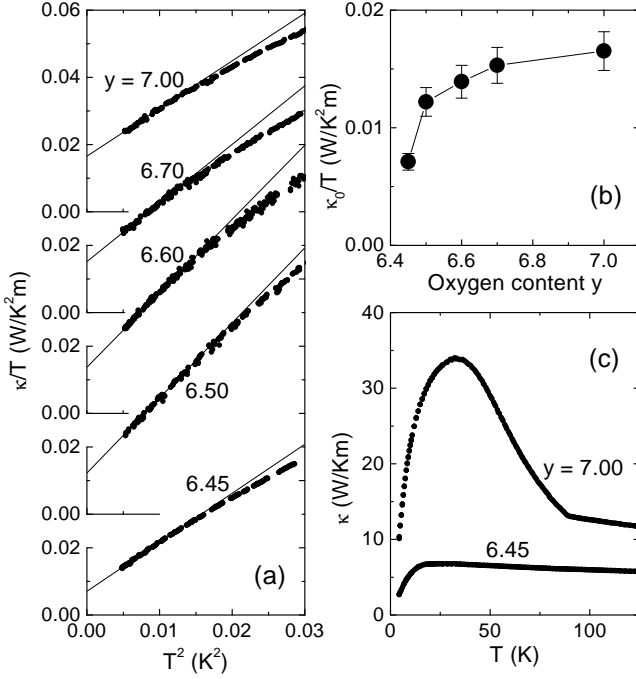


FIG. 1: (a) Temperature dependence of the in-plane thermal conductivity  $\kappa$  in 0 T for  $\text{YBa}_2\text{Cu}_3\text{O}_y$  single crystals with various  $y$  values. Thin solid lines are fits of the lowest- $T$  data to  $\kappa/T = a + bT^2$ . (b)  $y$  dependence of the residual QP component,  $\kappa_0/T$ . (c) High-temperature  $\kappa(T)$  data for  $y = 7.00$  and  $6.45$ .

$\text{Y}_2\text{O}_3$  crucibles by a conventional flux method. The crystals are carefully annealed to control the oxygen content and then perfectly detwinned [18]. Here we study untwinned single crystals with the oxygen content  $y$  of 6.45, 6.50, 6.60, 6.65, 6.70 and 7.00, for which the zero-resistivity  $T_c$ 's are 20, 39, 56, 59, 62 and 91 K, respectively. We emphasize that, to reliably control the oxygen content in the lightly doped regime, we have established elaborate procedures whose details are described in Ref. [19]. To probe the QP heat transport in the  $\text{CuO}_2$  planes, the thermal conductivity  $\kappa$  is measured along the  $a$  axis, with which the additional electronic heat transport coming from the Cu-O chains (that are along the  $b$  axis) can be avoided. Rectangular-shaped samples with a typical size of  $1.5 \times 0.5 \times 0.15 \text{ mm}^3$  are used for the measurements, where the longest (shortest) dimension is along the  $a$  ( $c$ ) axis. The measurements in the millikelvin region is done by a conventional steady-state “one heater, two thermometer” technique in a dilution refrigerator [20]. The magnetic-field dependence of  $\kappa$  is measured by the same method in a  $^3\text{He}$  refrigerator from 0.36 to 7 K. The magnetic field up to 16 T is always applied along the  $c$  axis. All the  $\kappa(H)$  data are taken with the field-cooled procedure [5, 21] to avoid the vortex-pinning-related hysteresis [22].

Figure 1(a) shows the thermal conductivity of the YBCO single crystals at very low temperatures down to

70 mK. At low enough temperature (below  $\sim 120$  mK), all the data are well described by  $\kappa/T = a + bT^2$  [see the thin solid lines in Fig. 1(a)], where the  $T$ -linear and cubic dependences of  $\kappa$  are attributed to the electron (or QP) term  $\kappa_e$  and the phonon term  $\kappa_{ph}$ , respectively [20, 23]. Thus, the zero-temperature intercept of the straight lines in Fig. 1(a) directly gives the value of the residual QP component,  $\kappa_0/T$ . This component  $\kappa_0/T$  is known to be due to the existence of extended zero-energy QP excitations near the nodes of the  $d$ -wave gap in the presence of even a small amount of disorder [24, 25], and is known to be “universal” against the change in the impurity concentrations [23]. Note that  $\kappa_0$  is a good approximation of  $\kappa_e$  only at very low  $T$ , since  $\kappa_e$  quickly grows with  $T$  [24, 26]. As shown in Fig. 1(b),  $\kappa_0/T$  increases gradually with carrier doping, which is similar to the behavior in LSCO [20] and is consistent with the previous YBCO result [27]. The smooth evolution of the residual QP thermal conductivity is compatible with a robust  $d$ -wave pairing symmetry in the whole doping range; namely, our data do not give any hint of a phase transition (as a function of  $y$ ) [28] that causes a change in the pairing symmetry and removes the nodes from the  $d_{x^2-y^2}$ -wave gap.

The high- $T$  thermal conductivity for the two limiting compositions of the present study,  $y = 7.00$  and  $6.45$ , are shown in Fig. 1(c). These data are taken by using a Chromel-Constantan thermocouple in a  $^4\text{He}$  cryostat. The peak value (34 W/Km) of our  $y = 7.00$  sample, which reflects the QP lifetime and is a measure of the cleanliness of the crystal [29], is considerably larger than the value ( $\sim 25$  W/Km) for ordinary YBCO crystals grown in YSZ crucibles [26, 29] and is close to the value ( $\sim 40$  W/Km) for the best crystals grown in  $\text{BaZrO}_3$  crucibles [26, 29]. This result demonstrates that our crystals are very clean, which is also evidenced in the electric transport data [18]. The  $y = 6.45$  sample shows little anomaly at  $T_c$  ( $= 20$  K), a behavior similar to that of underdoped LSCO [5].

The main result of the present work is shown in Fig. 2, where the magnetic-field dependences of  $\kappa$  is shown for all the underdoped samples. For optimally-doped (or slightly overdoped) YBCO, it is well known that  $\kappa$  is enhanced with  $H$  at low enough temperatures, but is suppressed with  $H$  at higher temperatures [26, 30]; such a behavior is consistent with the behavior of optimally-doped Bi2212 [21, 22] and LSCO [5]. Figures 2(c-e) demonstrate that such a behavior is also observed in underdoped samples with  $y = 6.60$ – $6.70$ ; from these data, one can conclude that there is essentially no qualitative change in  $\kappa(H)$  down to  $y = 6.60$ . However, upon further reducing the doping level, quite strong suppression in  $\kappa$  with increasing  $H$  suddenly shows up in the  $y = 6.50$  sample at 0.36 K [Fig. 2(b)]. The suppression of  $\kappa$  with  $H$  is also observed at  $y = 6.45$ . Therefore, there is a qualitative change in  $\kappa(H)$  at low temperature across  $y \simeq 6.55$ ,

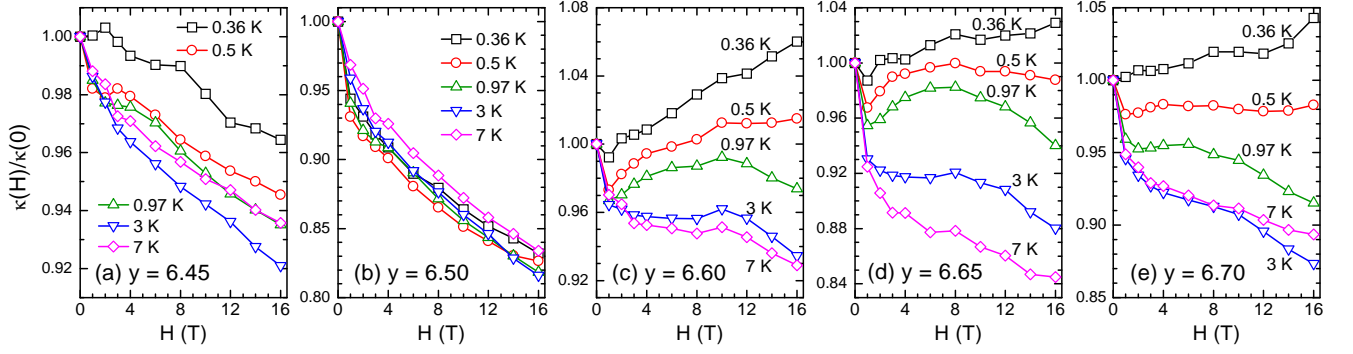


FIG. 2: Magnetic-field dependences of  $\kappa$  for underdoped YBCO single crystals. Note the distinct behavior at the lowest temperature for  $y \leq 6.50$  and for  $y \geq 6.60$ .

above which  $\kappa$  increases with  $H$  but below which it decreases with  $H$ . Note that the qualitative change in  $\kappa(H)$  is most certainly governed by the QP contribution to the heat transport, because it is unlikely that the *magnetic-field dependence* of the other contributions (phonons and magnetic excitations) suddenly change sign with doping. Note also that the smooth evolution of  $\kappa_0/T$  shown in Fig. 1(b) demonstrates that there is no sudden change in the pairing symmetry between  $y = 6.50$  and  $6.60$ , which means that the distinct change in the low- $T$   $\kappa(H)$  behavior is not due to a difference in the superconducting gap structure [21]. Thus, as in the case of LSCO [5, 6], the magnetic-field-induced enhancement or suppression of the QP heat transport at very low  $T$  reflects whether the electronic system is a thermal metal or a thermal insulator in high magnetic field, which in turn signifies whether the high-field normal state is a metal or an insulator. Hence, we can conclude that the MIC in YBCO occurs at  $y \simeq 6.55$ , which is deep in the underdoped regime and corresponds to  $p \simeq 0.07$  according to our systematic study of the Hall coefficient [19].

One may notice in Fig. 2(a) that the magnetic-field-induced suppression of  $\kappa$  becomes weaker with lowering temperature, which hints at the possibility that  $\kappa$  for  $y = 6.45$  might eventually increase with  $H$  at further lower temperature. To confirm that this is not the case, we have measured  $\kappa$  for  $y = 6.45$  in 16 T down to 100 mK and compared it to that in 0 T. As is shown in the inset to Fig. 3(a),  $\kappa(16\text{T})/\kappa(0\text{T})$  is always less than 1 and is declining with decreasing  $T$ , which assures that the magnetic-field-induced insulating state for  $y \leq 6.50$  concluded from Fig. 2 is indeed a zero-temperature property. The relative smallness of the  $H$  dependence in Fig. 2(a) is probably because the QP contribution to the total  $\kappa$  is the smallest in this most underdoped sample [see Fig. 1(b)].

It is useful to note that the data in the inset of Fig. 3(a) help us to estimate how high a magnetic field is needed to induce a true insulator with zero electrical conductivity: The data indicate that  $\kappa$  for  $y = 6.45$  is reduced by  $\sim 15\%$  in 16 T at 100 mK. At this temperature, the

fitting for  $y = 6.45$  in Fig. 1(a) tells us that  $\kappa$  is well described by  $\kappa/T = 7.32 \times 10^{-3} + 1.41T^2$ , where the first (second) term comes from  $\kappa_e$  ( $\kappa_{ph}$ ). Therefore, one can confidently estimate  $\kappa_e/\kappa$  to be  $\sim 0.34$  at 100 mK, and this estimate indicates that the 15% reduction of  $\kappa$  in 16 T is already halfway towards the complete suppression of  $\kappa_e$ ; this suggests that in the magnetic field of order 50 T the electronic contribution to  $\kappa$  would be quenched (*i.e.* a true insulator is achieved), which seems to be reasonable for a sample with  $T_c$  of 20 K [1].

Based on the above observations, one can draw a phase diagram of YBCO as depicted in the main panel of Fig. 3(a), where the MIC lies at  $y \simeq 6.55$ . Interestingly, in the light of the unusual doping-dependence of the in-plane resistivity anisotropy in YBCO [14], this value of the critical doping gives us a new insight into the nature of the MIC: As discussed in the introduction, one of the likely candidates for the cause of the QP localization observed in underdoped LSCO is the magnetic-field-induced stabilization of the charge stripes, if the static stripe state is competing with superconductivity [10]. In Ref. [14], it was demonstrated that in nearly optimally-doped YBCO the in-plane resistivity anisotropy ratio  $\rho_a/\rho_b$  has just a weak temperature dependence at high temperature and decreases at low temperature, which is probably dominated by the conductivity of the Cu-O chains that is expected to be diminished at low  $T$  (because of the strong tendency of the one-dimensional system to localization in the presence of disorder); on the other hand, at dopings lower than  $y \simeq 6.55$ ,  $\rho_a/\rho_b$  was found to *grow* with lowering temperature, which can be best attributed to the nematic charge stripes [10] globally along the  $b$  axis. In Fig. 3(b), we plot (using the data of Ref. [14]) the difference in  $\rho_a/\rho_b$  between 100 K and 300 K, which indicates whether the anisotropy grows or diminishes upon lowering temperature; when  $\rho_a/\rho_b(100\text{K}) - \rho_a/\rho_b(300\text{K})$  is positive, that means that the anisotropy grows with lowering  $T$  and thus is an indication that the stripes are becoming prominent in the charge transport. One can easily see that the boundary that separates the chain-dominated

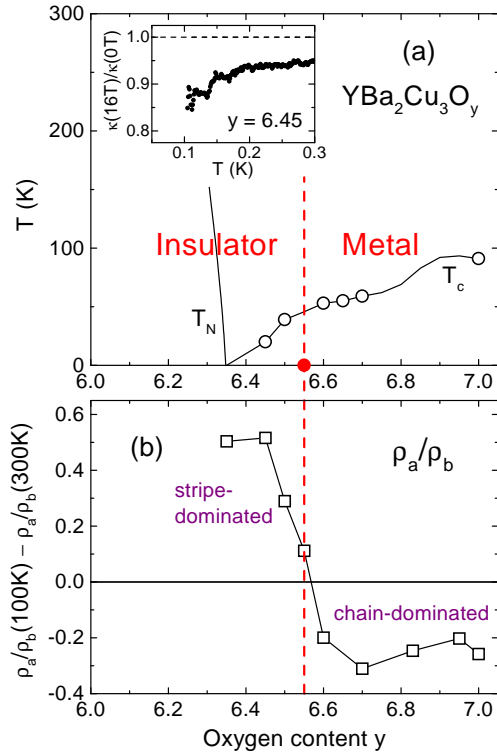


FIG. 3: (a) Phase diagram of YBCO.  $T_N$  and  $T_c$  indicate the Néel temperature [12] and the superconducting transition temperature [18], respectively. Open circles show  $T_c$ 's of the crystals studied in this work. The metal-insulator crossover in the normal state, determined from the low- $T$   $\kappa(H)$  behavior, occurs at  $y \simeq 6.55$  and is shown by a dashed line. Inset:  $T$ -dependence of the ratio  $\kappa(16T)/\kappa(0T)$  for  $y = 6.45$  down to 100 mK. (b) Difference between  $\rho_a/\rho_b$  at 100 K and that at 300 K; data are taken from Ref. [14].

and stripe-dominated regimes is located around  $y \simeq 6.55$  and this boundary corresponds exactly to the MIC determined from the  $\kappa(H)$  behavior.

This comparison naturally suggests that the low-doping side of the MIC (“insulator” regime) is where the stripe correlations are pronounced. In this regime, it is possible that the magnetic field can enhance the static ordering of the stripes (as was suggested by neutron [7, 8] and STM [9] measurements) and causes the electrons to be localized. It is useful to note that the critical doping for QPT in YBCO,  $p \simeq 0.07$ , is considerably lower than that in other cuprates; it is at  $p \simeq 1/8$  in BSLCO and at  $p \simeq 0.16$  in LSCO. This implies that, although the magnetic-field-induced static stripes are probably the common origin for the electron-localized state in the cuprates, the impact of the stripe correlations differs between the cuprates and such difference causes the variation in the location of the MIC.

In summary, we show that the magnetic-field dependence of the thermal conductivity of  $\text{YBa}_2\text{Cu}_3\text{O}_y$  single crystals measured at subkelvin temperatures indicates

that the metal-to-insulator crossover under high magnetic field takes place at  $y \simeq 6.55$ . Since the previous measurements of the in-plane resistivity anisotropy indicated [14] that the stripe correlations are pronounced in the transport below  $y \simeq 6.55$ , the present result gives strong support to the conjecture that existence of the stripe correlations characterizes the “insulating” normal state of the cuprates.

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